

Prediction of Underground Argon Content for Dark Matter Experiments

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In this paper, we demonstrate the use of physical models to evaluate the production of ^{39}Ar and ^{40}Ar underground. Considering both cosmogenic ^{39}Ar production and radiogenic ^{40}Ar production in situ and from external sources, we can derive the ratio of ^{39}Ar to ^{40}Ar in underground sources. We show for the first time that the ^{39}Ar production underground is dominated by stopping negative muon capture on ^{39}K and (α, n) induced subsequent $^{39}\text{K}(n, p)^{39}\text{Ar}$ reactions. The production of ^{39}Ar is shown as a function of depth. We demonstrate that argon depleted in ^{39}Ar can be obtained only if the depth of the underground resources is greater than 500 m.w.e. below the surface. Stopping negative muon capture on ^{39}K dominates over radiogenic production at depths of less than 2000 m.w.e., and that production by muon-induced neutrons is subdominant at any depth. The depletion factor depends strongly on both radioactivity level and potassium content in the rock. We measure the radioactivity concentration and potassium concentration in the rock for a potential site of an underground argon source in South Dakota. Depending on the probability of ^{39}Ar and ^{40}Ar produced underground being dissolved in the water, the upper limit of the concentration of ^{39}Ar in the underground water at this site is estimated to be in a range of a factor of 1.6 to 155 less than the ^{39}Ar concentration in the atmosphere. The calculation tools presented in this paper are also critical to the dating method with ^{39}Ar .

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I. INTRODUCTION

Argon is expected to be an excellent target for direct dark matter detection experiments searching for Weakly Interacting Massive Particles (WIMPs) [1, 2]. However, natural argon collected from the atmosphere contains ^{39}Ar and ^{42}Ar that are radioactive isotopes. The ratio of ^{39}Ar to ^{40}Ar is measured to be $8.1 \times 10^{-16} \text{g/g}$ [3–5] for atmospheric argon. Such a concentration results in 1 Bq/kg of specific activity of ^{39}Ar ($Q = 565 \text{ keV}$, $t_{1/2} = 269 \text{ y}$ [6]). Existing experimental limits on the ^{42}Ar concentration in the atmosphere are less than 10^{-18} ^{42}Ar atoms per ^{40}Ar atom [7–9], which makes about 0.018 Hz/kg of specific activity of ^{42}Ar ($Q = 3520 \text{ keV}$ and $t_{1/2} = 33 \text{ y}$). The production of ^{39}Ar in the atmosphere is dominated by the $^{40}\text{Ar}(n, 2n)$ process [3] with a reaction threshold of 1011.86 keV. ^{42}Ar is produced in the atmosphere through $^{40}\text{Ar} + n \rightarrow ^{41}\text{Ar}$ and $^{41}\text{Ar} + n \rightarrow ^{42}\text{Ar}$. The neutrons in the above reactions are from cosmic rays.

Recently developed argon-based dark matter detectors measure scintillation light induced by low-energy nuclear recoils due to elastic scattering of WIMPs. Argon has two distinct mechanisms for the emission of scintillation light: prompt light (6 ns) and delayed light ($1.5 \mu\text{s}$) [10]. The ratio of the prompt light to total light has been demonstrated experimentally to be well separated between elec-

tronic recoils and nuclear recoils [11, 12]. This property allows these two types of events to be distinguished on an event by event basis. The discrimination power is proved to be up to 10^7 electronic recoils versus one nuclear recoil [11, 12] at 20 keV electron-equivalent (ee) energy. Such a high pulse shape discrimination (PSD) power makes argon a very attractive dark matter target for the planned dark matter program at DUSEL. However, the existence of ^{39}Ar and ^{42}Ar potentially limits the detection sensitivity of next generation DUSEL dark matter experiments with multi-ton target masses. For instance, argon extracted from air results in ^{39}Ar beta-decay at a level of 1 Hz/kg [5]; a 5-ton detector would give a trigger rate of 5000 Hz and a total of 3.2×10^{10} events/year in the energy range of interest (20 keVee to 60 keVee); this requires PSD to be 3.2×10^{10} in order to achieve a sensitivity of $3 \times 10^{-46} \text{ cm}^2$ at 20 keVee threshold. Such a high PSD has not been yet demonstrated experimentally. Moreover, without depletion of ^{39}Ar , it is unlikely that the data acquisition dead time would be manageable for a multi-ton detector and a large scale computing facility would be required to perform on-line data reduction for a large amount of data from ^{39}Ar decays even for a single-phase detector. In addition, a dual-phase detector that drifts electrons from the liquid to gaseous would encounter serious difficulties with pile-up events because of the milliseconds drift time required. With depletion of ^{39}Ar by a large factor we can not only reduce background data, but also make it possible to lower the energy threshold to access a larger parameter space of WIMPs. Therefore, a multi-ton argon-based dark matter detector is promising only if natural argon

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is depleted of ^{39}Ar .

A large amount of argon depleted in ^{39}Ar and ^{42}Ar would be of great benefit to the DUSEL dark matter program described above. Because ^{39}Ar and ^{42}Ar are produced by cosmic rays in the atmosphere, it is logical to consider that underground sources of argon should be depleted in ^{39}Ar and ^{42}Ar due to the suppression of cosmic rays flux by the overburden. There are two sources of underground argon: 1) argon in natural gas and 2) argon dissolved in underground water. The former from natural gas wells in the US National Helium Reserve was found to contain a low level of ^{39}Ar [13]. The ratio of ^{39}Ar to ^{40}Ar (stable isotope) was found to be $\leq 4 \times 10^{-17}$ at a confidence level of 84% [13]. The latter from underground water 3200 feet below the surface at Wall, South Dakota is the focus of this paper.

Understanding the production of various argon isotopes underground by stopping negative muon captures and neutron interactions is critical to the dating method with ^{39}Ar . In this study, we realize for the first time that the ^{39}Ar production via stopping negative muon capture on ^{39}K is a dominant processes when the depth of underground resources is smaller than 1800 m.w.e. The early work of references [3, 14] showed the ^{39}Ar production only through (n,p) reaction on ^{39}K . Neutrons can be produced by: 1) cosmic-rays depending on the rock composition and depth of the site [15]; and 2) (α , n) neutrons depending on the rock composition and radioactivity level in the rock. The neutron fluxes and energy spectra are essential to the calculation of various isotopes of argon and hence the reliability of dating results. In this paper, we describe the production mechanisms underground for argon content in Section II. The measured rock composition and potassium content in a site as a potential argon source is discussed in Section III. We show the production rate of ^{39}Ar and ^{40}Ar in Section IV. Finally, we summarize our conclusions in Section V.

II. THE PRODUCTION MECHANISMS OF ARGON ISOTOPES UNDERGROUND

A. Production of ^{39}Ar

Underground production of ^{39}Ar isotopes occurs primarily through stopping negative muon captures and (n,p) reactions on ^{39}K and ^{40}Ca targets. Both elements are in a range of a few percent depending on the chemical composition of the rock. Neutrons are produced by three sources: 1) (α ,n) reactions induced by ^{238}U and ^{232}Th decay chains; 2) fission decays induced by ^{238}U and ^{232}Th ; and 3) muon-induced neutrons. The production of ^{39}Ar through stopping negative muon captures and muon-induced neutrons depends strongly on the depth of the rock overburden. This results in a depth dependent production of ^{39}Ar induced by stopping negative muons and muon-induced neutrons.

1. Stopping negative muon capture on ^{39}K

When negatively charged muons are stopped in the rock, they either decay through $\mu^- \rightarrow e^- \bar{\nu}_e \nu_\mu$ or are captured through $\mu(A, Z) \rightarrow \nu_\mu(A, Z-1)$. The capture cross section can be estimated using the formula

$$\sigma = \frac{A}{N_a v \tau \rho}, \quad (1)$$

where A is atomic mass number of the isotope, $N_a = 6.022 \times 10^{23} \text{ mol}^{-1}$ is Avogadro's constant, v is the speed of the stopping negative muons in the medium, τ is the stopping negative muon capture time in the medium, and ρ is the density of the medium. The lifetime of stopping negative muons in potassium is 410 ns [16]. The capture time can be calculated based on

$$\frac{1}{\tau} = \frac{1}{\tau_m} - \frac{1}{\tau_0}, \quad (2)$$

where τ_m (410 ns) is the measured lifetime of muons in potassium and τ_0 (2200 ns) is the lifetime of muons in vacuum. Using Eqs. (1) and (2), we obtain the cross section of the stopping negative muon capture on potassium as a function of the stopping muon energy.

The production rate of ^{39}Ar per year per gram of rock can be estimated using

$$P_1 = \frac{\rho}{A} \cdot N_a \cdot \phi \cdot \sigma \cdot f_{neg} \cdot \eta, \quad (3)$$

where P_1 is the production of ^{39}Ar per year per gram of rock, ρ is the density of potassium in the rock, ϕ is the stopping muon flux, σ is the capture cross section, $f_{neg} = 0.44$ is the fraction of negative muons [17], and η is the branching ratio of the yield of reaction $^{39}\text{K}(\mu^-, \nu)^{39}\text{Ar}$.

2. Neutron-induced ^{39}Ar production

In the rock formed thousand years ago, the rates of production and decay of ^{39}Ar should be equal. The production rate per year per gram of rock can be calculated by

$$P_2 = \phi_n \frac{N_K \sigma_{^{39}\text{K}(n,p)}}{N_{tot} \sigma_{abs}}, \quad (4)$$

where ϕ_n is the neutron flux in neutrons per year per gram of rock, N_K and N_{tot} are the number of potassium atoms and the total number of atoms in a gram of rock, and $\sigma_{^{39}\text{K}(n,p)}$ and σ_{abs} are the exothermic reaction $^{39}\text{K}(n,p)^{39}\text{Ar}$ cross section and neutron absorption cross section. Note that the neutron absorption here means that the neutron is absorbed and no further neutron is ejected, thus the absorption cross section includes contributions from all reaction channels which do not eject neutrons.

We calculate reaction cross sections by using the nuclear reaction code TALYS [18] version 0.72. The default

input parameters are used in this work. Fig. 1 shows the cross section of $^{39}\text{K}(n,p)$ reaction as a function of neutron kinetic energy. Also shown in the figure is the cross section of $^{42}\text{Ca}(n,\alpha)$ reaction, which can in principle contribute to ^{39}Ar production. However, its contribution is negligible because of relatively small cross section and ^{42}Ca concentration compared to ^{39}K .

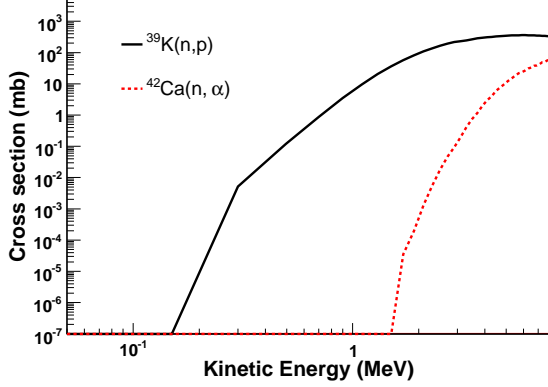


FIG. 1: ^{39}Ar production cross section as a function of neutron kinetic energy.

We plot in Fig. 2 the neutron absorption cross section of major atomic components in the rock. The black line illustrates the total neutron absorption cross section that is the weighted average, which uses the number of atoms as the weight factor.

The number of ^{39}Ar atoms is given by

$$N_{^{39}\text{Ar}} = \frac{P_1 + P_2}{\lambda}, \quad (5)$$

where P_1 and P_2 are the ^{39}Ar production rates given by Eq.(3) and Eq.(4), $\lambda = \ln 2/t_{1/2}$ is the decay constant of ^{39}Ar . The half-life of ^{39}Ar is 269 years.

B. The production rate of ^{40}Ar

^{40}Ar in rock is produced mainly by the decay of ^{40}K through electron capture with a branch ratio of 10.72%. The number of ^{40}Ar atoms produced can be estimated according to

$$N = N_0 B(1 - \exp(-\lambda t)), \quad (6)$$

where N_0 is original ^{40}K atoms at time $t = 0$, B is the branch ratio of electron capture, and $\lambda = \ln 2/t_{1/2}$ is the decay constant. The half-life $t_{1/2}$ of ^{40}K is 1.277×10^9 years. Fig. 3 shows the number of ^{40}Ar atoms produced as a function of the rock age. Although the natural abundance of ^{40}K is only 0.0117%, the number of ^{40}Ar produced in 65 million years reaches $\sim 1.7 \times 10^{14}$ atoms per gram of rock assuming a 2.59% of potassium content in the rock.

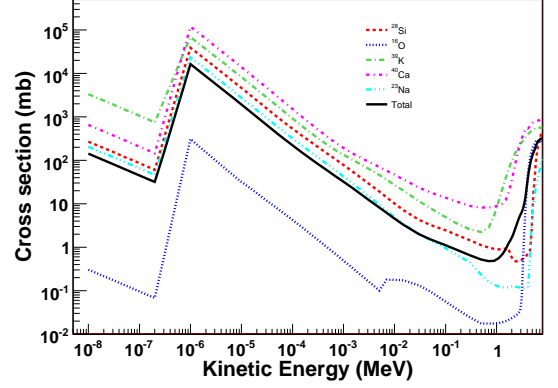


FIG. 2: The cross section of neutron absorption in different nuclei as a function of neutron kinetic energy. The black line shows neutron absorption cross section in the rock.

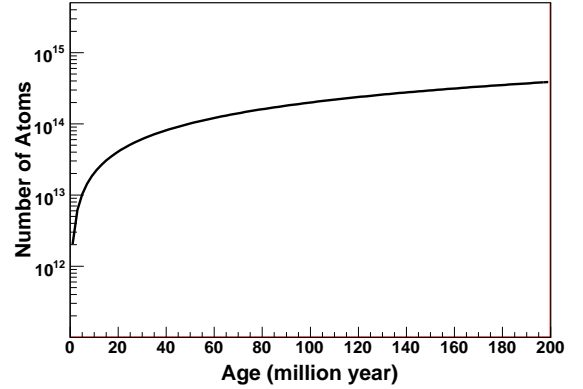


FIG. 3: The number of ^{40}Ar atoms produced in a gram of rock as function of the rock age.

Stopping negative muon capture on ^{40}Ca produces ^{40}K , which generates ^{40}Ar through electron capture decay. Because the production rate is much smaller than the existing ^{40}K in the rock, we ignore this process in our calculation of the ratio of ^{39}Ar to ^{40}Ar .

C. Calculation of the $^{39}\text{Ar}/^{40}\text{Ar}$ ratio as a function of depth

We utilize the fluxes of stopping negative muons and muon-induced neutrons as a function of depth derived in Ref. [15] to calculate the production of argon content with equations (3,4,5,6). The neutron energy spectrum induced by radioactivity obtained in Ref. [19] is used in the calculation.

The common elements found in the Earth's rocks are used in the calculation [20]. The eight most common elements [20] used in the calculation are listed in Ta-

ble I. The natural radioactivity concentrations of ^{238}U and ^{232}Th depend on the rock types. We assume the common rock in Table I is granite. Correspondingly, the radioactivity level of 4.7 ppm and 2.0 ppm of ^{238}U and ^{232}Th are used in the calculation [21]. Using the method described in Ref. [19], we obtain a yield of 5.5 n/(g·y). Subsequently, the ^{39}Ar production rate is estimated to be 7×10^{-3} atoms per year per gram of rock. The total number of ^{39}Ar atoms is 2.7 per gram of rock, which is independent of depth.

TABLE I: A list of the eight most common elements found in the Earth's rocks.

Elements	Chemical Symbol	Mass Weight (%)
Oxygen	O	46.60
Silicon	Si	27.72
Aluminum	Al	8.14
Iron	Fe	5.00
Calcium	Ca	3.63
Sodium	Na	2.83
Potassium	K	2.59
Magnesium	Mg	2.09

The depth dependence of the ^{39}Ar production is shown in Figure 4. Note that the rock chemical composition varies from location to location. However, the dependence of the production of ^{39}Ar due to the change in the rock chemical composition is much weaker than that of the change in depth.

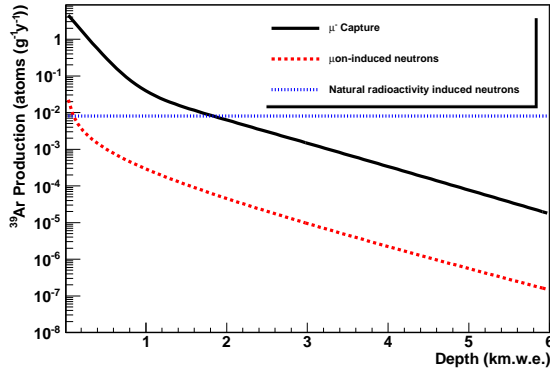


FIG. 4: The ^{39}Ar production as a function of depth. The eight common elements found in the Earth's rocks are used in the calculation.

If one assumes that ^{39}Ar and ^{40}Ar have the same possibility to diffuse out of the grains of the rock and to be mixed with existing gas in the pore volume, then the $^{39}\text{Ar}/^{40}\text{Ar}$ ratio in the pore volume can be expressed as

$$R = \frac{N_{^{39}\text{Ar}} \times p}{N_{^{40}\text{Ar}}^0 + N_{^{40}\text{Ar}}^d \times p} \quad (7)$$

where p is the possibility of ^{39}Ar and ^{40}Ar being mixed in the pore volume, $N_{^{40}\text{Ar}}^0$ is the original number of ^{40}Ar

contained in a certain amount of gas due to the ^{40}Ar concentration in ambient air, which is related to the pore volume, $N_{^{40}\text{Ar}}^d$ is the number of ^{40}Ar atoms produced underground through ^{40}K EC decays. Using Eq. (7), the ratio of the ^{39}Ar to ^{40}Ar can be calculated with known natural abundance of ^{39}K in the rock. Given a rock density of 2.7 g/cm³, a rock porosity of 20%, and an assumption that the pore volume is filled with gas with the ambient air concentrations (78%/21%/1% of N₂/O₂/Ar), the ratio of ^{39}Ar to ^{40}Ar can be determined. The results are shown in Figure 5.

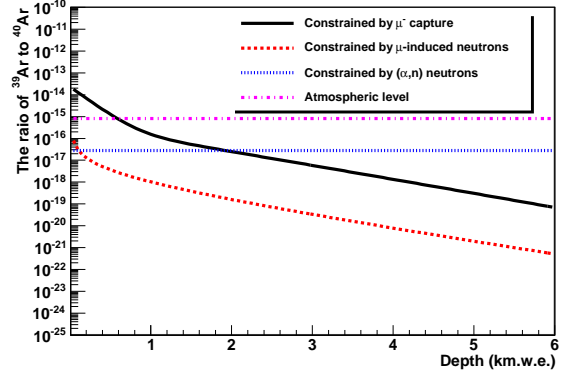


FIG. 5: The ratio of the ^{39}Ar to ^{40}Ar calculated using formulas and methods described in text. The eight common elements found in the Earth's rocks are used in the calculation. Shown is a ratio assuming that the produced argon is mixed at a 10% level by underground gas.

Eq.(7) can be applied to a underground water reservoir, where p is the possibility of ^{39}Ar and ^{40}Ar being dissolved in water. For a rock density of 2.7 g/cm³, a rock porosity of 20%, and an assumption that the pore water contains gas with the ambient air concentrations (78%/21%/1% of N₂/O₂/Ar) at a solubility of 61 mg/L for argon, the ratio of ^{39}Ar to ^{40}Ar as a function of depth is shown in Figure 6.

As can be seen in Fig. 4, the larger the depth, the smaller the production of ^{39}Ar . On the other hand, at the large depth, the production of ^{39}Ar is dominated by stopping negative muon capture and the neutrons from natural radioactivity. Since $^{40}\text{Ca}(n, 2p)^{39}\text{Ar}$ reaction requires neutron energy to be greater than 8.3 MeV and the majority of the (α, n) neutrons induced by radioactivity is less than this reaction threshold, the production of ^{39}Ar at large depth underground is dominated by stopping negative muon capture on ^{39}K and $^{39}\text{K}(n, p)^{39}\text{Ar}$. Hence, the understanding of ^{39}K content and radioactivity level in the rock surrounding the gas field or water reservoir where the argon gas is extracted from is essential.

The underground production of ^{40}Ar is primarily from ^{40}K via electron capture decays. Therefore, an accurate knowledge of the potassium content is critical. The underground production of ^{42}Ar is negligible.

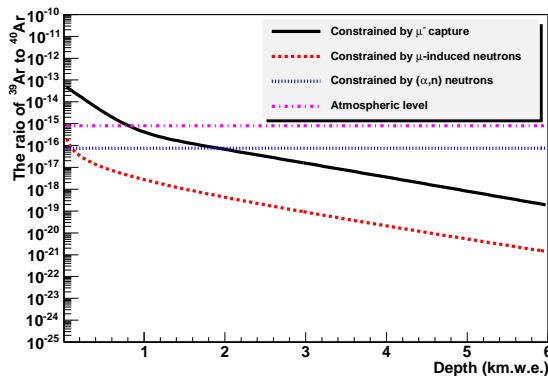


FIG. 6: The ratio of the ^{39}Ar to ^{40}Ar calculated using formulas and methods described in text. The eight common elements found in the Earth's rocks are used in the calculation. Shown is a ratio assuming that the produced argon is dissolved at a 10% level by underground water.

III. EXPERIMENTAL RESULTS OF POTASSIUM CONTENT

A. Underground water Site

The community of Wall, South Dakota pumps water using seven wells that extract water from the Fall River aquifer in an old rock formation 3200 ft below the surface. The average water consumption is 200 gallons per minute. The water temperature underground is about 42°C . The expected argon gas extraction is about 2.3 kg per day assuming 70% efficiency with 61 mg/L solubility. However, the level of ^{39}Ar in argon gas extracted from well water is contingent upon the understanding of the rock composition especially the potassium content in the rock. Two different samples obtained from the South Dakota Geological Survey were evaluated. The first sample was a cutting sample from the 3200 ft level in the well that was provided by the well drilling company. The second sample was a core sample taken from the Fall River Aquifer at a distance of about 40 miles from Wall.

B. Method

The samples were broken down using a mortar and pestle and then passed through a 100 mesh sieve. The samples were then dried in an oven set at 100°C to remove the water. After drying the samples were consistently stored in a desiccator to prevent re-hydration. Initially the amount of free K (K not in the soil matrix) was found using the following solution preparation and utilizing the atomic absorption spectrometer. The solution preparation consisted of 2 g of the sample, 40 ml of water (Note that all water used was nanopure water that had been purified to 18 M Ω cm with a SYBRON/Barnsted Nanopure System), 15.5 ml of concentrated hydrochloric

acid (HCl), and 2 ml of concentrated nitric acid (HNO_3). One important observation was that the solution did not boil when the acids were added indicating a lack of carbonates. The solution was heated with stirring, and finally filtered to remove the remaining soil particles. The filtered solution was placed in a 250 ml volumetric flask and diluted with water to the 250 ml mark. An atomic absorption analysis was done on this sample using a Thermo Jarrell Ash atomic absorption spectrometer which contained a Smith-Hieftje 11 spectrophotometer. The light source was a hollow cathode lamp produced by the Thermo Jarrell Ash Corporation which emitted at 766.5 nm. The bandpass was set at 1.0 nm and the lamp current was set to 5 mA. Two runs were done with the instrument for each sample. The measurement of the total K in the soil samples was measured under the assumption that most of the K was tied up in a SiO_2 compound. This was concluded from the X-ray diffraction analysis conducted on the sample. The sample preparation was obtained from Buck Scientific [22]. One gram of the soil sample was mixed with 5 grams of a 2:1 flux of lithium carbonate (LiCO_3) and zinc oxide (ZnO). The sample and flux were placed in a platinum crucible and fused in a muffle furnace that was heated to 950°C for 20 minutes. The melt was then allowed to cool and was re-dissolved using 20 ml of HCl and 20 ml of water. This solution was filtered to remove the soil particles that had not been re-dissolved. A series of standards that ranged from 0.01 ppm to 5.0 ppm were prepared and analyzed with the atomic absorption spectrophotometer yielding a calibration curve of $y = (0.0562 \pm 0.0009)x - (0.0034 \pm 0.0021)$ with a correlation coefficient of 0.9994. Potassium samples were diluted to fall within the calibration curve. Once again two runs were done and then averaged. Three different samples from the core sample and from the cutting sample were treated in this manner. Finally, we obtain a potassium content of 18500 ± 300 ppm.

IV. PRODUCTION RATE OF ARGON ISOTOPES

In this particular site in South Dakota at 3200-ft below the surface, the stopping muon flux is calculated to be $3.4 \times 10^{-8} \text{ cm}^{-2} \text{ s}^{-1}$. Utilizing Eq.(3), we obtain a ^{39}Ar production of 0.0026 per year per gram of rock.

The natural radioactivity of ^{232}Th and ^{238}U is analyzed to be around 0.6 ppm and 1.9 ppm, separately. The rock is sandstone with a chemical composition described in Table II.

The (α, n) neutron yield in the rock is calculated [19] to be 0.25 n/(g·ppm·y) for ^{232}Th and 0.65 n/(g·ppm·y) for ^{238}U . The fission decay of ^{238}U induced neutron yield is 0.52 n/(g·ppm·y). The total neutron yield induced by radioactivity is 2.4 n/(g·y). Using Eq.(4), the ^{39}Ar production rate is estimated to be $\sim 3 \times 10^{-3}$ atoms per year per gram of rock.

The ^{39}Ar produced by the muon-induced neutrons at

TABLE II: The chemical composition of the sandstone 3200 ft below the surface at Wall, South Dakota.

Elements	Mass Weight (%)
Si	43.0
O	50.71
K	1.85
Fe	1.5
Kr	1.0
Na	0.7
Mg	0.6
Al	0.5
Ca	0.14

this depth is negligible. The total number of ^{39}Ar atoms given by Eq.(5) is estimated to be 2.17 atoms per gram of rock.

If we assume that ^{39}Ar and ^{40}Ar have the same possibility to diffuse out of the grains of the rock and to be dissolved in the pore water, then the $^{39}\text{Ar}/^{40}\text{Ar}$ ratio in water can be obtained by applying Eq. (7),

$$R = \frac{2.17p}{N_{Ar} + 1.2 \times 10^{14}p} \quad (8)$$

where p is the possibility of ^{39}Ar and ^{40}Ar being dissolved in water, and N_{Ar} is the original number of ^{40}Ar contained in a certain amount of water, which is related to the pore volume. If we assume that the porosity is 20% (this is typical for sandstone [3]), then the water volume corresponding to a gram of rock with the measured density of 2.2 g/cm^3 is $9.1 \times 10^{-2} \text{ cm}^3$. Given a total saturation solubility of 125 mg/L for $\text{N}_2/\text{O}_2/\text{Ar}$ at 20°C assuming the gas has ambient air concentrations (nitrogen/oxygen/argon of 78%/21%/1%) with different solubility of N_2 (20 mg/L)/ O_2 (44 mg/L)/ Ar (61 mg/L), the dissolved argon in water is $61 \text{ mg/L} \times 0.01 / (20 \text{ mg/L} \times 0.78 + 44 \text{ mg/L} \times 0.21 + 61 \text{ mg/L} \times 0.01)$. This results in a 2.4 % of argon content (3 mg/L) in the total gas content in the water. Therefore, $N_{Ar} = 4.1 \times 10^{15}$. The term of 1.2×10^{14} is the number of ^{40}Ar atoms from ^{40}K decay with a total concentration of 18500 ppm in a ~ 65 million-year rock formation. Fig. 7 shows the ratio of ^{39}Ar to ^{40}Ar atoms contained in water as a function of p . Thus, a depletion factor of 1.6 to 155 can be achieved for the argon extracted from this site when p varies from 0.01 to 1.0. Considering saturation solubility decreases when temperature increases, the dissolved possibility of ^{39}Ar and ^{40}Ar can be as small as 1% for the argon content produced in the underground environment where the temperature is higher. This would make underground water reservoir a valuable source for depleted argon that can be used for ton-scale dark matter experiments. If one assumes that 30% [3] of Ar produced will be dissolved in water, the ratio between ^{39}Ar and ^{40}Ar in water is $\sim 1.6 \times 10^{-17}$, which is a factor of ~ 5 lower than that in atmosphere.

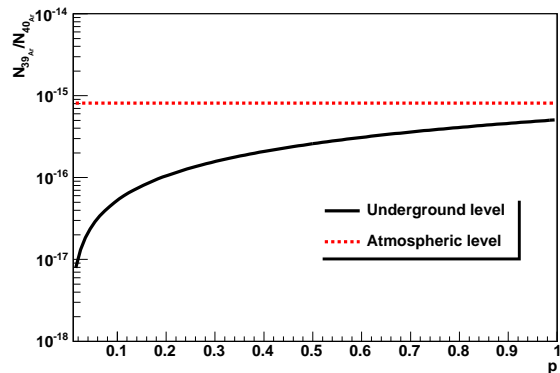


FIG. 7: The ratio of ^{39}Ar to ^{40}Ar atoms contained in water as a function of the dissolved probability (p).

V. CONCLUSION

We have evaluated that the ^{39}Ar production underground through (n, p) reactions induced by neutrons and stopping negative muon capture. We demonstrate that argon depleted in ^{39}Ar can be obtained only if the depth of the underground resources is greater than 500 m.w.e. below the surface. Stopping negative muon capture on ^{39}K dominates over radiogenic production at depths of less than 2000 m.w.e., and that production by muon-induced neutrons is subdominant at any depth. Depending on the rock porosity and argon diffusion coefficient, the upper limit of the concentration of ^{39}Ar in the underground water at Wall of South Dakota is estimated to be in a range of a factor of 1.6 to 155 less than the ^{39}Ar concentration in the atmosphere. Naturally occurring ^{40}K EC decay dominates the content of ^{40}Ar underground. The ratio of ^{39}Ar to ^{40}Ar depends largely on the following: 1) the potassium content in the rock; 2) the natural radioactivity of ^{232}Th and ^{238}U in the rock; 3) the rock porosity; 4) argon gas diffusion coefficient underground; and 5) argon gas solubility in the water underground. We estimate that the production of ^{42}Ar is negligible at this underground site. The calculation tools presented in this paper are also critical to the dating method with ^{39}Ar [3, 14, 23, 24]. We conclude that the ratio of ^{39}Ar to ^{40}Ar varies from location to location depending on the ^{39}K content and the radioactivity level in the rock at large depth. Extraction of argon gas from underground sources must: 1) evaluate the depth of the site, the age of the rock, the rock composition, and radioactivity; 2) study the porosity of the rock and argon gas diffusion coefficient; and 3) understand the solubility of argon gas as a function of pressure and temperature. The algorithm described in this paper is critical to understanding the discrepancy in the dating methods with the ratio of ^{39}Ar to ^{40}Ar [14, 25–27].

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